CASE PP/1-22413/P2/CGC 2073

CERTIFICATE OF MAILING

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8/6/03 — Date

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF
STEPHEN M. ANDREWS ET AL
APPLICATION NO: 10/034,048
FILED: DECEMBER 20, 2001
FOR: POLYESTER COMPOSITIONS OF LOW RESIDUAL ALDEHYDE CONTENT

Group Art Unit: 1712
Examiner: Patricia A. Short

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER RULE 132

I, Stephen M. Andrews, the undersigned, states:

That I was awarded a Ph.D. in Chemistry from Virginia Commonwealth University in 1987;

That I have approximately 16 years of chemical research and development experience; That from 1992 to date I have worked in the Research and Development laboratories of Ciba Specialty Chemical Corporation;

That I presently hold the position of Technical Fellow;

That the following experiments were performed by me or under my supervision:

That I am submitting herewith the following exact report of the tests made and the results obtained.

It was the object of the tests reported below to compare acetaldehyde reduction in polyethylene terephthalate substrates by using dipentaerythritol and polyacrylamide (MAP 1070, a blend of 75-80% polyacrylamide with 20-25% polyethylene glycol) alone and in combination.

The conditions for testing and analysis below are the similar as that for Application No. 10/034,048 above.

General - PET bottle grade pellets are subjected to extrusion compounding to simulate the heat history which PET experiences when thermally injection molded into bottle preforms and subsequently stretch-blow molded into bottles. The efficacy of an additive added to reduce the formation of acetaldehyde is determined by quantitative analysis using thermal desorption GC-MS or GC-FID after adoption of published methods. An unstabilized PET is extruded each day to provide a control polymer for measuring acetaldehyde formation.

Extrusion - PET is pre-dried in vacuo under nitrogen at an oven temperature of about 70°C to a moisture level of about 30 ppm which is verified on a Mitsubishi VA-O6 moisturemeter. A Leistritz 18 mm or 27 mm co-rotating, non-intermeshing twin screw extruder is configured as follows: set temps = throat (220-230°C), zones and die (270°C), actual extrudate melt temperature is 275-280°C, screw at 100-110 rpm, hopper feeder = 10-15 ppm.

Injection molding – a polyester is pre-dried in vacuo at an oven temperature about 70°C, to a suitably low moisture level. A BOY injection molding machine equipped with 60 mil, 2"x2" mold, is generally configured as follows: zone 1 =500F, zones2-nozzle =550F, inj. P = 500 psi, mold temp=80F, screw speed 125 rpm.

Acetaldehyde Analysis - The concentration of acetaldehyde in PET is quantitatively determined using a thermal desorption GC-FID method adapted from B. Nijassen et al., Packaging Technology and Science, 9, 175 (1996); S. Yong Lee, SPE ANTEC 1997, pp 857-861; and M. Dong et al., J. Chromatographic Science, 18, 242 (1980). A general example follows below:

The PET samples are analyzed, typically in triplicate, by weighing 500 mg of powdered PET pellets (obtained by cryogenically pulverizing ~ 20 grams of the PET) in a 5 mL crimp sealed headspace vial. The sample vial is heated at 120°C for one hour in a Tekmar model 7000 static headspace analyzer. The headspace gas (5 cc) is then transferred via a heated transfer line to a Varian 3600 GC-FID system for quantification of the acetaldehyde (AA). By using a known acetaldehyde value for PET, the ratio of peak areas for the known PET resin and for the experimental PET resin blends are compared and the amount of acetaldehyde in the experimental blend can be obtained.

The results are shown below for PET resin from Mitsui and Kosa

Kosa PET

Formulation **% Additive** **AA ppm** **% red'n AA**

	additive	GCFID	
	Kosa PET	**AA**	**% red'n**
	wgt %	**Ppm**	**AA**
neat pet	0	3.26	**n/a**
dipentaerythritol	0.40	2.78	14.7
dipentaerythritol alone	0.500	2.66	18.4
MAP 1070 alone	0.05	2.87	12.0
MAP 1070 alone	0.1	2.53	22.4
MAP 1070 alone	0.5	*interfering GC peaks*	
dipentaerythritol + MAP 1070	0.450 0.050	0.57	**82.5**
dipentaerythritol + MAP 1070	0.400 0.1000	1.09	**66.6**

Mitsui PET

Formulation % Additive AA ppm % red'n AA

	Additive	GCFID	
	Mitsui PET	**AA**	**% red'n**
	wgt %	**Ppm**	**AA**
Neat pet	0	3.35	n/a
Dipentaerythritol alone	0.25	2.72	18.8
Dipentaerythritol alone	0.3750	2.75	17.9
Dipentaerythritol alone	0.500	2.66	20.6
Dipentaerythritol + MAP 1070	.250 .2500	0.11	**96.7**
Dipentaerythritol + MAP 1070	.375 .125	0.42	**87.5**
MAP 1070 alone	0.1250	1.93	42.4
MAP 1070 alone	0.250	1.23	63.3

Conclusions

Note in the Mitsui PET substrate the calculated additive effect of acetaldehyde reduction for dipentaerythritol and MAP 1070 would be 82.1% and 60.3% whereas, the actual effect of combining the two additives gives a synergistic effect of 96.7% and 87.5% respectively. The same effect is also noted in the Kosa PET. The calculated additive effect for acetaldehyde reduction would be 37.1% and 26.7% whereas the actual effect is 66.6% and 82.5% respectively.

It is clear from these results that the co-use of dipentaerythritol in combination with MAP 1070 gives synergistic, surprising reduction in acetaldehyde residues in PET.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

6 Aug 2003

Date

Stephen M. Andrews